PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

REC'D 28 JUL 2005
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Applicant's or agent's file reference	FOR FURTHER ACTION	See Form PCT/IPEA/416
3194.1032-002		
International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/US2004/023594	22.07.2004	23.07.2003

International Patent Classification (IPC) or national classification and IPC
C01B3/04, C01B3/56, C01B3/02, C01C1/04, C25B1/04, C01B23/00, C01C1/02

Applicant
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1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 5 sheets, including this cover sheet.
3. This report is also accompanied by ANNEXES, comprising:
   a. ☒ *sent to the applicant and to the International Bureau)* a total of 5 sheets, as follows:
      ☒ sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).
      ☐ sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.
   b. ☐ *(sent to the International Bureau only)* a total of (indicate type and number of electronic carrier(s)) , containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).

4. This report contains indications relating to the following items:

☒	Box No. I	Basis of the opinion
☐	Box No. II	Priority
☐	Box No. III	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
☐	Box No. IV	Lack of unity of invention
☒	Box No. V	Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
☐	Box No. VI	Certain documents cited
☐	Box No. VII	Certain defects in the international application
☒	Box No. VIII	Certain observations on the international application

Date of submission of the demand	Date of completion of this report
23.05.2005	27.07.2005

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INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/US2004/023594

Box No. I Basis of the report

1. With regard to the **language**, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.

   ☐ This report is based on translations from the original language into the following language , which is the language of a translation furnished for the purposes of:
   - ☐ international search (under Rules 12.3 and 23.1(b))
   - ☐ publication of the international application (under Rule 12.4)
   - ☐ international preliminary examination (under Rules 55.2 and/or 55.3)

2. With regard to the **elements*** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report)*:

Description, Pages	
1-10	as originally filed
Claims, Numbers	
1-23	filed with telefax on 23.05.2005
Drawings, Sheets	
1/3-3/3	as originally filed

   ☐ a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing

3. ☐ The amendments have resulted in the cancellation of:
   - ☐ the description, pages
   - ☐ the claims, Nos.
   - ☐ the drawings, sheets/figs
   - ☐ the sequence listing *(specify)*:
   - ☐ any table(s) related to sequence listing *(specify)*:

4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).
   - ☐ the description, pages
   - ☐ the claims, Nos.
   - ☐ the drawings, sheets/figs
   - ☐ the sequence listing *(specify)*:
   - ☐ any table(s) related to sequence listing *(specify)*:

* *If item 4 applies, some or all of these sheets may be marked "superseded."*

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/US2004/023594

Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1-23
	No:	Claims	
Inventive step (IS)	Yes:	Claims	1-23
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-23
	No:	Claims	

2. Citations and explanations (Rule 70.7):

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Reference is made to the following documents:
D1: US-A-4 427 504 (STUCKI SAMUEL) 24 January 1984 (1984-01-24)
D2: US 2003/094098 A1 (WATANABE TADAHARU ET AL) 22 May 2003 (2003-05-22)

2. Document D1, which is considered to represent the most relevant state of the art, discloses (cf. col.3 l.47-col.4 l.20) the electrolysis of water for hydrogen production. Initially the water is treated to eliminate dissolved or suspended liquid, gaseous and/or solid substances. The hydrogen is then mixed with nitrogen from an air distillation unit to produce ammonia.
The subject-matter of independent claim 1 differs from D1 in that the ammonia is synthesized at a point of use and in that prior to ammonia synthesis the hydrogen and the nitrogen undergo a purification step.

The problem to be solved by the present invention may therefore be regarded as providing high purity ammonia for LEDs production.

The solution to this problem proposed in claim 1 of the present application is considered as involving an inventive step (Article 33(3) PCT) for the following reasons:
The document D2 (cf. paragraphs [0003]-[0007];[0011]-[0012];[0016];[0028];[0045]) relates to the field of purification of gases used in the semiconductor industry, in particular nitrogen, hydrogen and ammonia. The purifier material comprises oxides of e.g. molybdenum, tungsten, manganese, iron, lithium, sodium, potassium, titanium.
Although document D2 is concerned with the same problem as the present application and recognises the same drawbacks in delivering ultra-high purity gases, it proposes a different solution, i.e. to purify delivered gases at the point of use.
There is no mention in this document to the production of high-purity ammonia at a point of use.

The applicant describes several advantages of point of use production of ammonia: the inventive method "eliminates the contamination associated with delivery from bulk storage or cylinders and the need for layers of distributed purification" (p.5 l.26-27), avoids the

problem of heavily contaminated fluids at the bottom of a delivery cylinder (p.3 l.15-17) and the need for a distribution system for purified gases (p.5 l.29-p.6 l.3).

The same argumentation applies mutatis mutandis to the subject-matter of independent claims 21, 22 and 23.

Hence, the subject-matter of the independent claims is considered novel and involves an inventive step (Article 33(1)-(3) PCT).

3. Claims 2-20 are dependent on claim 1 and as such also meet the requirements of the PCT with respect to novelty and inventive step.

4. The subject-matter of the claims is industrial applicable (Article 33(4) PCT).

Re Item VIII

Certain observations on the international application

Although claims 1, 21 and 23 have been drafted as separate independent claims, they appear to relate effectively to the same subject-matter and to differ from each other only with regard to the definition of the subject-matter for which protection is sought. The aforementioned claims therefore lack conciseness and as such do not meet the requirements of Article 6 PCT.

CLAIMS

What is claimed is:

1. A method for producing ammonia at a point of use from liquid water and nitrogen, comprising:

feeding a quantity of de-ionized water to a hydrogen generator;

producing a quantity of hydrogen from the quantity of de-ionized water utilizing said hydrogen generator;

producing a quantity of purified hydrogen by passing said quantity of hydrogen through a hydrogen purifier;

producing a quantity of purified nitrogen by passing a quantity of nitrogen through a nitrogen purifier; and

contacting said quantity of purified hydrogen and said quantity of purified nitrogen with a catalyst bed, wherein a portion of said purified hydrogen and a portion of said purified nitrogen react to form a quantity of ammonia at the point of use.

2. The method as recited in claim 1, further comprising:

de-gassing said quantity of de-ionized water prior to feeding the de-ionized water to said hydrogen generator, to remove a portion of dissolved gasses in said quantity of de-ionized water.

3. The method as recited in claim 2, wherein said quantity of de-ionized water is de-gassed in a membrane contactor, having a first stage followed by a second stage.

4. The method as recited in claim 3, wherein in said first stage, a first portion of said dissolved gassed are removed by nitrogen stripping.

5. The method as recited in claim 3, wherein in said second stage, a second portion of said dissolved gasses are removed by vacuum stripping.

6. The method as recited in claim 1, further comprising:

compressing said quantity of purified hydrogen and said quantity of purified nitrogen prior to contacting said quantity of purified hydrogen and said quantity of purified nitrogen with said catalyst bed.

7. The method as recited in claim 6, wherein said quantity of purified hydrogen and said quantity of purified nitrogen are compressed to a pressure between about 10 to about 100 atmospheres, absolute.

8. The method as recited in claim 1, further comprising:

producing a quantity of purified ammonia at the point of use by passing said quantity of ammonia through an ammonia purifier.

9. The method as recited in claim 8, further comprising:

delivering a portion of said quantity of purified ammonia to a semiconductor process tool at the point of use.

10. The method as recited in claim 8, wherein said ammonia purifier comprises a high surface area metal oxide comprising oxides of barium, calcium, iron, lithium, manganese, molybdenum, potassium, rhenium, sodium, strontium, titanium, tungsten or vanadium.

11. The method as recited in claim 8, wherein at least one of said ammonia purifier, said hydrogen purifier, and said nitrogen purifier are regenerated with a portion of said quantity of purified hydrogen.

12. The method as recited in claim 8, wherein the concentration of an impurity in said quantity of purified ammonia is reduced to less than about 50 ppb.

13. The method as recited in claim 8, wherein the concentration of an impurity in said quantity of purified ammonia is reduced to less than about 10 ppb.

14. The method as recited in claim 1, wherein said hydrogen purifier comprises

a high surface area metal oxide comprising oxides of barium, calcium, iron, lithium, manganese, molybdenum, nickel, potassium, rhenium, sodium, strontium, titanium, tungsten, or vanadium; and
optionally, metallic nickel.

15. The method as recited in claim 1, wherein said nitrogen purifier comprises a nickel catalyst.

16. The method as recited in claim 1, wherein said hydrogen generator produces hydrogen from water by electrolytic means.

17. The method as recited in claim 1, wherein the concentration of an impurity in said quantity of purified hydrogen is reduced to less than about 50 ppb.

18. The method as recited in claim 1, wherein the concentration of an impurity in said quantity of purified nitrogen is reduced to less than about 50 ppb.

19. The method as recited in claim 1, wherein the concentration of an impurity in said quantity of purified hydrogen is reduced to less than about 10 ppb.

20. The method as recited in claim 1, wherein the concentration of an impurity in said quantity of purified nitrogen is reduced to less than about 10 ppb.

21. A method for producing point of use ammonia from liquid water and nitrogen, comprising:

de-gassing a quantity of de-ionized water, to remove a portion of dissolved gasses in said quantity of de-ionized water;

feeding a quantity of said de-ionized, de-gassed water to a hydrogen generator;

producing a quantity of hydrogen from the quantity of said de-ionized, de-gassed water utilizing said hydrogen generator;

producing a quantity of purified hydrogen by passing said quantity of hydrogen through a hydrogen purifier;

producing a quantity of purified nitrogen by passing a quantity of nitrogen through a nitrogen purifier;

compressing said quantity of purified hydrogen and said quantity of purified nitrogen;

contacting said compressed quantity of purified hydrogen and said compressed quantity of purified nitrogen with a catalyst bed, wherein a portion of said purified hydrogen and a portion of said purified nitrogen react to form a quantity of ammonia;

producing a quantity of purified ammonia at the point of use by passing said quantity of ammonia through an ammonia purifier; and

delivering a portion of said quantity of purified ammonia to a semiconductor process tool at the point of use.

22. A method for producing purified hydrogen gas from liquid water, at a point of use comprising:

feeding a quantity of de-ionized water to a degasser sufficient to remove oxygen to a level of about 1 ppb or less and CO_2 to a level of about 1 ppm or less;

feeding said degassed water to a hydrogen generator;

producing a quantity of hydrogen from the quantity of de-ionized water utilizing said hydrogen generator;

producing a quantity of purified hydrogen at the point of use by passing said quantity of hydrogen through a hydrogen purifier.

23. A method for producing ammonia at a point of use from liquid water and nitrogen, comprising:

feeding a quantity of de-ionized water to a hydrogen generator;

producing a quantity of hydrogen from the quantity of de-ionized water utilizing said hydrogen generator;

producing a quantity of purified hydrogen by passing said quantity of hydrogen through a hydrogen purifier;

producing a quantity of purified nitrogen by passing a quantity of nitrogen through a nitrogen purifier; and

contacting said quantity of purified hydrogen and said quantity of purified nitrogen with a catalyst bed, wherein a portion of said purified hydrogen and a portion of said purified nitrogen react to form a quantity of ammonia at the point of use; and

producing a quantity of purified ammonia at the point of use by passing said quantity of ammonia through an ammonia purifier.